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Cerenkov Type Phase-matched Second Harmonic Generation in Polymeric Channel Waveguides

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Fabrication of polymer channel waveguides by etching on glass is reported. Results on second harmonic generation (SHG) in the Cerenkov mode from stable poled polymeric channel waveguides are presented. The waveguide conversion efficiency for doubling of Nd:YAG laser beam was estimated to be 0.1%/W with a total efficiency of 4.9 X 10^{-7}%/W.

Nonlinear optical (NLO) polymers have been investigated extensively because of their potential applications. These materials offer many advantages over inorganic materials and organic crystals, such as easy processing, high optical damage threshold and low cost. In order to exhibit second order NLO properties, polymers have to be poled to induce the noncentrosymmetric structure [1]. Unfortunately in most of the NLO polymers, the NLO chromophores tend to randomize with time. Recently a photocrosslinking technique has been utilized to maintain the alignment of the NLO molecules against thermal relaxation for poled polymers. Crosslinking of the aligned molecules is carried out by irradiating the polymer with UV light during the late stage of poling [2-5].

Waveguide Second Harmonic Generation (SHG) has recently attracted considerable interest because it is a promising way to obtain inexpensive, efficient, short wavelength coherent sources [6]. In earlier reports NLO polymeric films have been utilized for waveguide electro-optic modulatiors [7,8]. Relatively few studies on phase-matched waveguide SHG have been reported since the precise control of phase-matching condition is difficult in waveguides Among various different approaches [6,9-10] in waveguides for frequency doubling, Cerenkov type phase-matched method [11] is attractive because of its simplicity. The Cerenkov phase matching condition is not very stringent and the NLO polymeric materials can be even somewhat absorbing at the doubled frequency. Sugihara et al. recently reported Cerenkov SHG from a copolymer of methyl methacrylate and Disperse Red 1 (DR1) substituted methylacrylate [10]. However, these materials have a small value of d_{33} and they exhibit some relaxation of the second-order nonlinearity. Stable nonlinear response and large values of nonlinearity are both important factors in achieving superior device performance.

Earlier Cerenkov type phase matched second harmonic generation from poled crosslinked polymeric planar waveguides was reported by us [12]. In this paper, we report frequency doubling results in the Cerenkov mode from crosslinked polymer channel waveguides. Frequency doubled green light was observed and the waveguide conversion efficiency was found to be high. A simple method for the fabrication of channel waveguides is also presented.

A channel waveguide was fabricated on glass substrates using standard photolithographic techniques. A positive photoresist (Microposit S1813) was spin-coated on a cleaned glass substrate (at coating speed of 4000 RPM). The photoresist coated substrate was placed in an oven and baked for 30 min at 100^0C. After baking, a mask that defined the channels was put on top of the photoresist layer on the substrate, and then the photoresist was exposed for 10 seconds to UV light (peak at 365 nm). The photoresist was developed for 50 seconds. The glass slide was baked again for 30 min at 100^0C. Buffered Oxide Etch was then used to etch the exposed substrate region. Typical etching time is usually about 4 min. The groove depth was 0.95 μm for a 4 min etching time. These values were determined using a Surface Profiler. The etched waveguide channel width for the waveguide was approximately 5 μm. Fig. 1 summarizes the processing steps for the channel waveguide fabrication. The surface of the etched glass slide was observed under an optical microscope and was found to be of good optical quality.

The structure of the DGEBA-NAC polymer is shown in Fig. 2. The polymer is an epoxy of Diglycidyl Ether of Bisphenol A and 4-nitroaniline functionalized with cinnamoyl groups (hence DGEBA-NAC). Photocrosslinking occurs through 2+2

cycloaddition. The detailed synthesis of this polymer has been reported earlier [3]. Fig. 3 describes the absorption spectrum of DGEBA-NAC in tetrahydrofuran (THF).

Films were prepared on channeled and thick end-polished glass substrates by spin-coating (spin speed 600 RPM) a solution of the polymer in a propylene glycol methyl ether acetate (PGMEA) solvent (weight ratio 1:4). The samples were placed in a vacuum oven at 40^0C for 12 hours to remove residual solvent. The refractive indices of the polymer and of the substrate were measured by an ellipsometer and are summarized in Table 1. The film thickness was estimated to be 1.4 μm from the interference pattern in the transmission spectrum.

The sample was slowly heated up to a temperature around the glass transition temperature (T_g) (83^0C). The samples were poled by corona discharge in a wire-to-plane geometry. After 5 min of poling, UV light was irradiated for 5 min to crosslink the poled polymer with the poling field on. The source of UV light is a mercury lamp producing an intensity of 1 mW/cm^2 with emission peak at 254 nm on the polymer surface. After exposure, the temperature of the sample was lowered to room temperature with the poling field on.

The channel waveguide frequency doubling experimental set up is shown schematically in Fig. 4. The polymer channel waveguide consisted of the channelled glass substrate, the NLO polymeric film overlayer and air. TM waves were selected by a polarizer. TM waves have the largest nonlinear interactions with NLO material when they travel in the waveguide since d_{33} is the largest second-order NLO coefficient in a poled polymer waveguide. The light source was a Q-switched Nd:YAG laser with a pulse width of 200 ns and a repetition rate

of 12.9 kHz. A microscope objective (40 X) was used to focus the incident light beam and end-fire couple light into the channel waveguide. The fundamental beam emerging from the waveguide edge was blocked by filters ($CuSO_4$ and interference filters) in order to measure the SH intensity. A CCD camera system was set up to monitor the coupling and confinement of light in the waveguide which was easily observed with the monitor.

According to the dispersion equation [13], the fundamental TM wave phase-matching condition requires that DGEBA-NAC film thickness be between 1.29 and 1.55 μm at the fundamental wavelength of 1.064 μm. This requirement is not very stringent and easily fulfilled by spin coating techniques. The SH peak output power, corrected for the filter loss, was 3.31 μW when the incident peak power was 26 W and was found to be proportional to the square of the fundamental power (Fig. 5). The waveguide conversion efficiency was calculated to be 0.1%/W for an input fundamental power of 53.4 mW and a waveguide interaction length of 1.1 cm. Thus the total conversion efficiency normalized by the fundamental power was found to be 4.9 X 10^{-7}%/W due mostly to the losses in coupling the fundamental into the waveguide from the YAG laser. This waveguide conversion efficiency is much larger than those reported earlier [10,12], mainly due to large fundamental beam intensity, confinement in a channel and the low SH absorption by the NLO polymer used in the present work. The waveguide loss was measured to be 2.4 dB/cm.

Channel waveguides were successfully fabricated on glass substrates using standard photolithographic techniques. Frequency doubling experiments were performed using these stable poled polymer waveguides. The waveguide conversion efficiency was 0.1 %/W with a total efficiency of 4.9 X 10^{-7}%/W. By

increasing the coupling efficiency into the guide with the use of fibers and a modified waveguide design we expect that the net total efficiency can be considerably improved. Although quantitative measurements were not carried out, a bright green light was observed even when a cw Nd: YAG laser was used.

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Figure Captions

Fig. 1. Processing steps of the channel waveguide fabrication. (a) photoresist coating, (b) patterned exposure, (c) development, (d) etching, (e) channeled substrate.

Fig. 2. The structure of the DGEBA-NAC polymer.

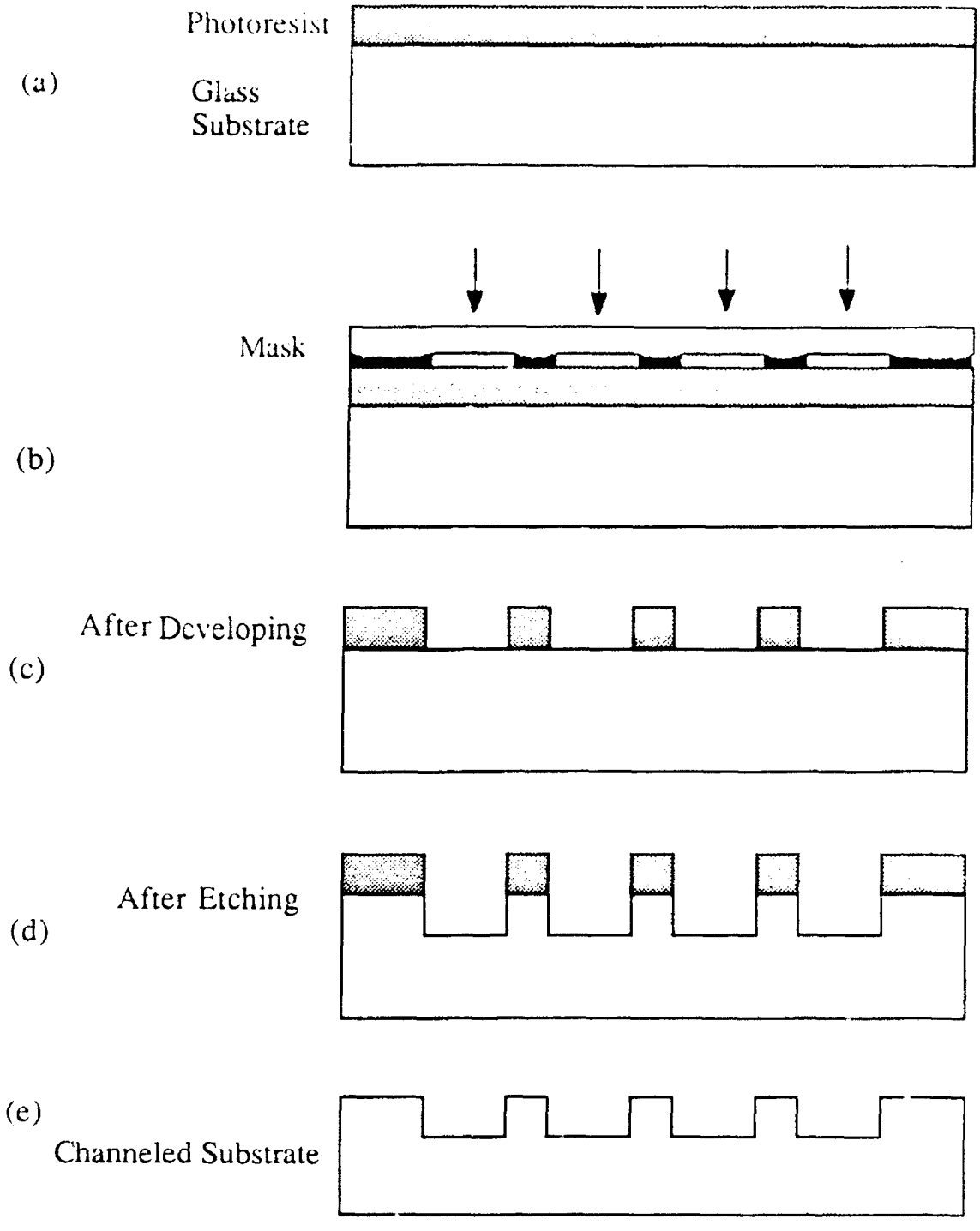
Fig. 3. Absorption spectrum of the DGEBA-NAC in THF.

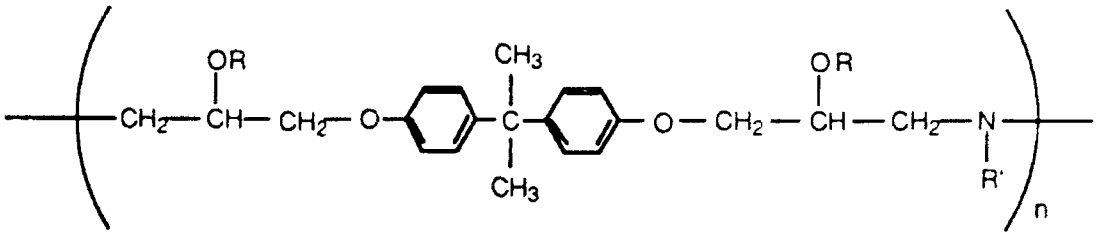
Fig. 4. Top view of channel waveguide frequency doubler.

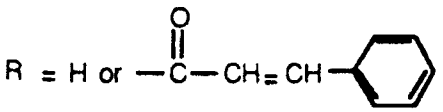
Fig. 5. Second harmonic intensity versus fundamental intensity on logarithmic scales.

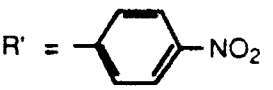
Table I Properties of the NLO polymer and the glass substrate.

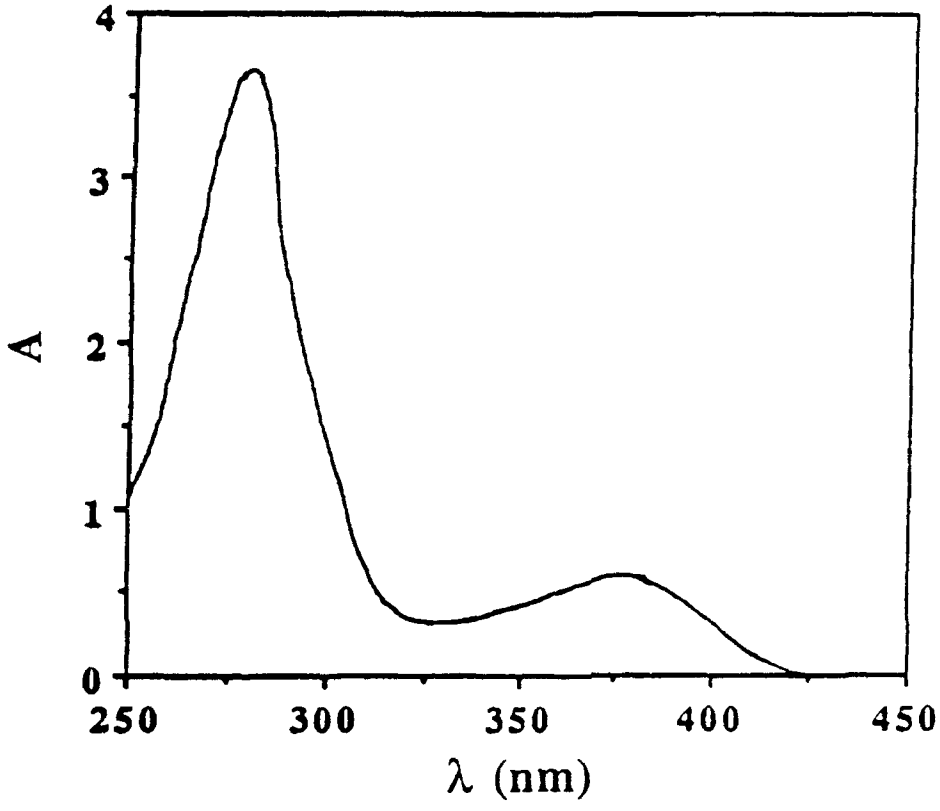
	DGEBA-NAC	Glass
n at λ = 532 nm	1.637	1.522
633 nm	1.635	1.514
1000 nm	1.613	1.502
Waveguide loss (dB/cm)	2.4 (λ = 1.064 μm)	
d_{33} (pm/V) (λ = 1.064 μm)	8.1	


Photoresist
(a)
Glass
Substrate
Mask
(b)
After Developing
(c)
After Etching
(d)
(e)
Channeled Substrate








4
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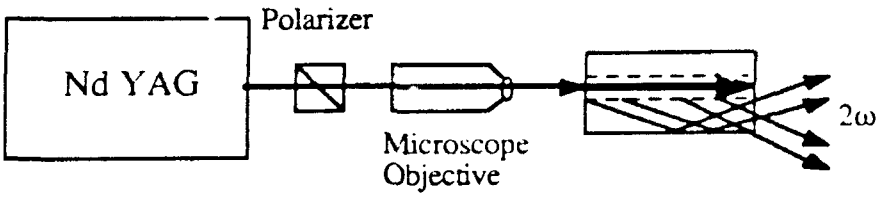
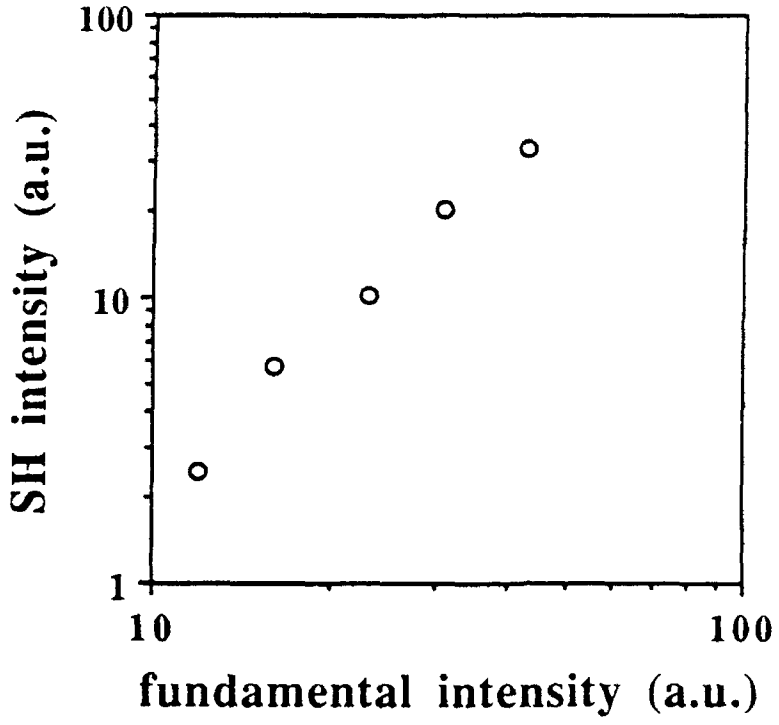

Polarizer
Nd YAG
Microscope
Objective
2ω


100
10
1
SH intensity (a.u.)
10
100
fundamental intensity (a.u.)